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Research paper

Betavoltaic Battery using Platinum/Porous ZnO Schottky Junction

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Abstract

Background and Objectives: Semiconductor junction-based radioisotope detectors are commonly used in radioisotope batteries due to their small size and excellent performance. This study aims to design a betavoltaic battery based on a metal-porous semiconductor Schottky structure, comprising an N-type zinc oxide (ZnO) semiconductor and platinum (Pt) metal.

Methods: we utilized the TCAD-SILVACO 3D simulator to simulate the device, and a C-Interpreter code was applied to simulate the beta particle source, which was an electron beam with an average energy equivalent to 63Ni beta particles. The short circuit current, open-circuit voltage, fill factor (FF), and efficiency of the designed structure were calculated through simulation. Additionally, we discussed the theoretical justification based on the energy band structure.

Results: The energy conversion efficiency of the proposed structure was calculated to be 11.37% when bulk ZnO was utilized in the Schottky junction. However, by creating pores and increasing the effective junction area, a conversion efficiency of 35.5% was achieved. The proposed structure exhibited a short-circuit current, open-circuit voltage, and fill factor (FF) of 37.5 nA, 1.237 V, and 76.5%, respectively.

Conclusion: This study explored a betavoltaic device with a porous structure based on a Schottky junction between Pt and ZnO semiconductor. The creation of pores increased the contact surface area and effectively trapped beta beams, resulting in improved performance metrics such as efficiency, short circuit current, and open-circuit voltage.

Introduction

There are several methods for converting the energy of decayed particles into electrical energy. Using semiconductor devices to convert the energy of decayed beta particles into electrical energy is among the practical methods in this field.

Due to the rapid progress in microelectromechanical systems (MEMS) and nanoelectronics, there is a great demand for nuclear batteries [1]. Popularity of these batteries is rooted in their commercial, military, and medical applications, which are viable alternatives for chemical batteries due to their longevity, especially in remote applications. nuclear batteries, consisting of a radioactive source and a semiconductor junction or photonic crystals, have received great attentions in many works due to their stable output and longevity [2]-[7]. The PN structure is a widely used semiconductor structure that has a depletion region in the junction section. This region, with an appropriate width, absorbs Beta particles and produces electron-hole pairs. To improve the efficiency and output power of betavoltaic cells, various structures such as p-n, p-i-n, and Schottky have been studied and analyzed in references [8]-[13]. As the depletion region increases, the interaction of beta particles with the semiconductor material increases, resulting in the production of more electron-hole pairs.

The PIN structure, with a wider depletion zone, increases the probability of beta absorption [14]-[19]. The Schottky structure junction is created between the semiconductor and the metal near the surface. The Schottky diode has higher radiation resistance than that of the p–n junction and is a promising converter of the betavoltaic battery [20]-[23]. When determining the structure, it is important to consider the diffusion length of beta particles, which varies depending on the source type, activity, half-life, and particle type such as beta or alpha. Beta particles cause less damage to the cell structure and are commonly used in betavoltaic cells. Promethium-147 (147PM) is used as a beta source for designing betavoltaic batteries in references [24], [26]. Rosenkranz et al. presented the Clinical experience with nuclear-powered pacemakers (Promethium-147) in references [25]. The radioisotope 35S is used for designing and optimizing Si-35S betavoltaic liquid nuclear batteries [27]-[29]. Tritium (3H)-powered betavoltaic cells are presented in references [30]-[32]. The 63Ni betavoltaic battery is a common battery presented in different research studies [33]-[36]. Researchers have conducted evaluations of 90Sr betavoltaic power sources [37]-[39]. The 14C is also a radioisotope that can be used as a C-betavoltaic energy converter [40], but it has complicated production methods [41]-[42].

To increase the energy conversion efficiency of the betavoltaic cells, different methods are applied. Generally, the cell material and the cell structure are two important parameters. From material point of view, using wide-bandgap materials in betavoltaic cells have improved the cell characteristics. Wide-bandgap semiconductors have high radiation hardness and increase the energy conversion efficiency of the cell due to the raise of the open-circuit voltage of the cell [43].

From structure point of view, it should be noted that a significant amount of the energy from the decayed particles is lost in flat structures and bulk junctions due to the angular distribution of beta particles as well as low effective junction area. It is possible to use nanowires and porous structures at p-n, p-i-n and Schottky junctions to greatly increase the effective area of the junction and the propagated electric fields. In this regard, various designs and structures have been investigated [44]. In Si-based betavoltaic devices, using porous Si increases efficiency by 0.22% [45]. Creating nanowire structures in gallium phosphide improves the cell efficiency compared to the bulk material [46]; for example, the efficiency of a betavoltaic microbattery based on TiO2 nanotubes with 63Ni source and activity of 8 mCi is reported to be 7.3% [47]. theoretical support for the parameter analysis of betavoltaic batteries is presented in [48] and is used for study on the series resistance of betavoltaic batteries.

Despite many efforts conducted and different methods presented to improve the betacell characteristics, there is still a need for an efficient betavoltaic cell to simultaneously benefits from optimum structure and material.

Schottky diode has many applications for radiation detectors. An InSb Schottky detector, fabricated from an undoped InSb wafer with Hall mobility was used for alpha particle detection. The output pulse of this InSb detector showed a very fast rise time, which was comparable with the output pulses of scintillation detectors [49].

In this study, a betavoltaic device is designed based on a Schottky junction between Pt metal and ZnO semiconductor with a porous structure. The significant difference in work function between Pt (5.64 eV) and ZnO [50] results in an effective Schottky barrier. In the proposed device, ZnO serves as a wide energy bandgap material with high energy hardness; The porosity of ZnO effectively increase the junction area, leading to a higher number of trapped beta particles. To achieve the desired beta-generated electron-hole concentration inside the structure, empirical dose functions are simulated in C++ code and the results are linked to SILVACO-TCAD 3D simulator. The radioisotope 63Ni with an activity of 1 mCi, due to its long half-life, is employed in the C++ code as the source of beta particles. The structure of the device and Schottky barrier formed at the Pt-ZnO junction are thoroughly investigated. Subsequently, the results obtained from the SILVACO simulator are presented. Finally, an analysis is conducted on the results of the porous structure for different metals and compared with other relevant samples.

Device Structure

The structure was simulated using the TCAD-SILVACO 3D simulator. A Schottky junction was formed by employing an n-type ZnO semiconductor and Pt metal. The Pt layer was extended on the surface of the ZnO structure, and zinc oxide nanowires were applied to the irradiated surface. ZnO nanowires can be created through the porosity of the ZnO surface, which increases the effective cell area and enhances the electric fields between pores [51]. This effectively improves the trapping of emitted beta particles in the cells and increases the contact surface area. The trapped particles generate electron-hole pairs in ZnO, which are then collected by the internal electric field of the Schottky junction, thereby improving efficiency (refer to the simulation and results section). The porous surface consists of comb-shaped indentations with a height of 2 µm. Table 1 presents the structure dimensions. To reduce beta particle flux attenuation, a small thickness is considered for the anode electrode, ranging from 0.04 to 0.1 µm. The anode electrode in the upper part is exposed on the processed porous surfaces of zinc oxide. The cathode electrode is created on the backside of the ZnO wafer, which also serves as the substrate for the structure in the simulations. ZnO nanowires are considered on the surface. (see Fig. 1 for a 3D schematic of the structure).

ZnO is a material with wide energy bandgap and excellent properties. It has high band gap energy (3.37 eV) [52] and can be used as a suitable material in piezoelectric converters, optical waveguides, acoustic wave devices, varistors, sensors, etc. In addition to its widespread use in current industries, ZnO is considered as a “material of the future” [53]. ZnO crystallizes in Wurtzite (B4 type) structure at the ambient pressure and temperature. This structure is a hexagonal lattice belonging to the space group P63mc and is characterized by two interconnected subnets of Zn^{2+} and O^{2-}. This regular tetrahedral arrangement creates polar symmetry which is an important factor in crystal growth and etching [50]. In betavoltaic devices, semiconductor structure should be exposed to radiation for a long time. Therefore, the used material should have good resistance to radiation damage so as not to cause rapid damage to the device [54]-[56]. ZnO semiconductor exhibits excellent hardness against radiation damage, so that it is not damaged by high-energy electron bombardment (> 1.6 Mev) [57]. It can be simply doped to achieve an n-type ZnO semiconductor [58]-[59]. In this work, ZnO was doped with 10^{15} atom/cm^3 to achieve the n-type semiconductor.

Table 1: Structure dimensions

Device length	5.9 um
Device width	3.8 um
Device height	6 um
Length of ZnO Nanowires	0.7 um
width of ZnO Nanowires	0.4 um
height of ZnO Nanowires	2 um

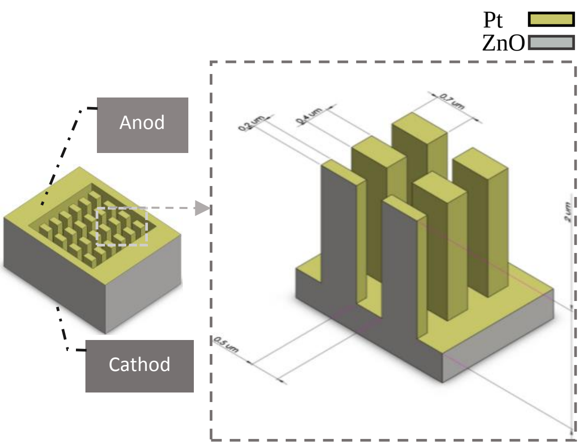

Fig. 1: The 3D schematic of the proposed betacell. The structure is a Schottky junction between ZnO and Pt.

A. Metal-Semiconductor Junction

Most of the useful properties of p-n junction can be achieved simply by forming a suitable junction between the metal and semiconductor. When a metal with work function ϕ_m is connected to a semiconductor with work function ϕ_s ($\phi_s<\phi_m$), a depletion region is formed in the semiconductor near the junction by transferring free electrons from the semiconductor to the metal and an electric field is created in this region to separate electron-hole pairs made by radiation [60] (Fig. 2). Schottky junction between Pt and ZnO in betavoltaic cell. Pt has Schottky contact with ZnO. The depletion region of the junction is hashed. Beta particles are incident at Pt and generate electron-hole pairs in ZnO. The beta-generated carriers are then separated in depletion region of the junction and can generate current and voltage in the external circuit. The energy band diagram of the junction is also presented on top of the junction schematic. The Schottky barrier potential of the junction and contact potential are demonstrated as qϕB and qϕi in Fig. 2. Schottky junctions are superior to p-n junction in terms of being used in radioisotope devices due to various reasons, including their simpler fabrication mechanism.

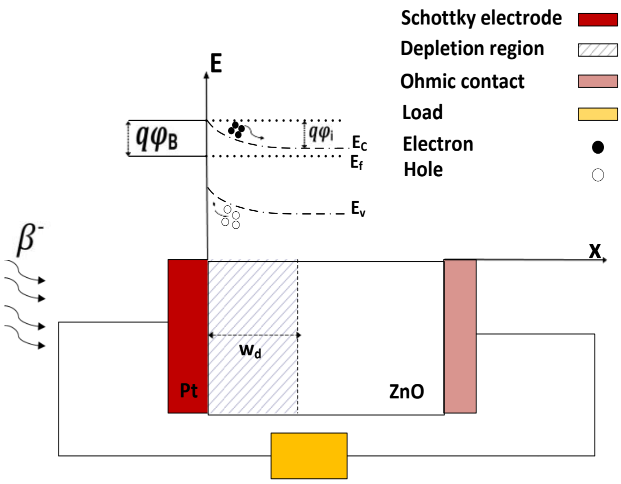

Fig. 2: Schottky junction between Pt and ZnO and The energy band diagram of the junction.

The Schottky junction barrier is defined by the metal work function and semiconductor electron affinity, as presented by (1).

$$\varphi_B = \varphi_m - X \quad (1)$$

where X is ZnO electron affinity (4.1- 4.5 eV) [50], [61]-[62] and ϕ_B is Schottky barrier height, which is 1.54 eV in Pt/ZnO junction. Figs. 3(a) and 3(b) demonstrate current-voltage curve in dark mode and energy band diagram of the structure, respectively, showing formation of Schottky junction and barrier at ZnO-Pt junction. The I-V curve of Pt/porous ZnO in dark mode; The rectifying behavior of the curve demonstrates that the Schottky barrier is created at the interface of metal and semiconductor (Fig 3(a)). Energy band diagram of the junction between Pt and ZnO. As shown, a potential barrier with the height of 1.54eV is created at the ZnO surface (Figs. 3(b)). Table 2 presents density and work function of some metals that can be used to form Schottky or ohmic junctions with ZnO. Metals with large work function are suitable for forming Schottky junction with ZnO. Hence, Pt, Ir, Ni, Pd, and Au metals were examined. The results presented in the comparison section showed Pt was suitable for forming Schottky junction with ZnO.

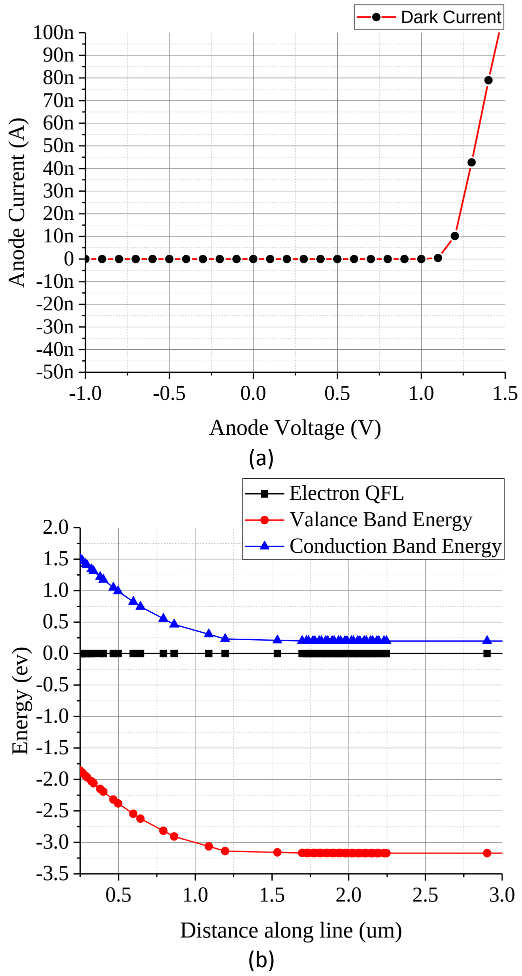

Fig. 3: a) The I-V curve of Pt/porous ZnO in dark mode. b) Energy band diagram of the junction between Pt and ZnO.

Table 2: Work function and density of some metals for forming junction with ZnO [50]

Metal	φm	Density ($\frac{gr}{cm^3}$)
Ta	4.25	16.69
Ag	4.26	10.5
Al	4.28	2.7
Ti	4.33	4.51
Zn	4.47	7.133
Mo	4.6	10.28
Au	5.1	19.32
Pd	5.12	12.02
Ni	5.15	8.9
Ir	5.25	22.56
Pt	5.64	21.45

B. Radioisotope source

Radioisotopes have received great attention due to their high energy storage. They release this energy during their half-life. Long half-life is among the important factors for the sources of betavoltaic devices. In general, radioisotope is selected based on the type of radiation, energy, specific activity, cost, and half-life.

In this work, ^{63}Ni beta source with the half-life of 100.2 years, average energy of 17.4 keV, and maximum energy of 67 keV is simulated by C-Interpreter code method. According to (2), the radiant power of 10^{-7} W is obtained for the activity of 1 mCi.

$$P_{Ni-63} = 3.7 \times 10^{10} \times A \times q \times E_{avg} \quad (2)$$

where A stands for the source activity, E_{avg} is the average energy of radioisotope source and q is the electron charge.

Simulations and Results

The presented Pt/porous ZnO Schottky junction is tested under an electron beam with the average energy of ^{63}Ni beta particles. To achieve the beta-generated carrier concentrations inside the device, an analytical function for electron-hole generation rate is simulated in C-interpreter code and linked to SILVACO (Fig. 4).

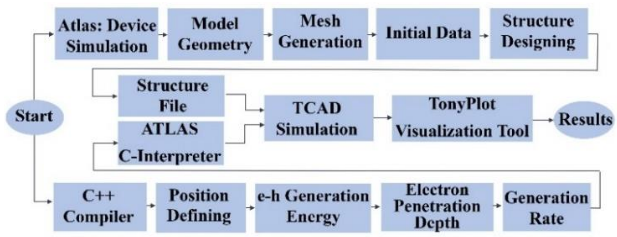

Fig. 4: Simulation flowchart.

As it is known, after the design of the structure in the ATLAS section of the SILVACO software, the C++ code is entered into the simulation as a source of beta radiation by using one of the SILVACO code abilities (the F.RADIATE BEAM statement). In this simulation, E. Yakimov's analytical function [63] is used to express the electron-hole generation rate, G(r) by (3):

$$G(r) = G_0 F(r, y, E) h(y, E) \quad (3)$$

In this equation, G_0, $F(r,y,E)$, and $h(y,E)$ refer to lateral-dose function, and depth-dose function respectively. The local rate of the electron-hole pair generation, G_0, depends on the electron beam energy(E), electron beam current(I), and the energy required to generate an electron-hole pair in material (e_i) as (4):

$$G_0 = \frac{EI}{qe_i} \quad (4)$$

in which q is the electron charge. The quantity e_i can be obtained as (5) [64]:

$$G_0 = \frac{EI}{qe_i} \quad (5)$$

e_i=2.596E_g + 0.714

Eg refers to the bandgap of the material, that is equal to 3.37 ev for zinc oxide. The depth-dose function, h(y, E), is calculated using Everhart [65] expression as (6):

$$h(y,E) = 0.6 + 6.21 \times R_{norm} - 12.4 \times R_{norm}{}^2 + 5.69 \times R_{norm}{}^3 \quad (6)$$

R_{norm} is the depth; more accurately it is the depth (y) normalized by the penetration depth of the beam R(E), which can be calculated as (7) [66]:

$$R(E) = \frac{0.0276 \times A \times E^n}{Z^{0.89} \times \rho} \quad (7)$$

where A is the atomic weight(g/mol), E is the beam energy (Kev), ρ is the density, and Z is the atomic number. n is often chosen to be 1.67 when E>5 Kev.

For the lateral-dose function, F(r,y,E), the empirical expression as proposed by Donolato by (8) is used [69], [74], [75]:

$$F(r,y,E) = \frac{1.76}{2\pi\sigma^2 R(E)}\exp[-(\frac{r}{\sigma})2] \quad (8)$$

where, σ^2 = 0.36δ2 + 0.11$\frac{y^3}{R(E)}$, δ is the electron beam diameter, r^2=x^2 and δ = 0.01×R(E).

The above-mentioned functions are used in C++ code to calculate the distribution of generated electron-hole pairs in the device.

Efficiency is among the important parameters of energy converters which is defined according to (9):

$$\eta = \frac{P_{out}}{P_{total}} \quad (9)$$

where P_{out} is the output power of betavoltaic device, which can be obtained by (10) and P_{total} is the total power received from the radioisotope source, which is equal to 10^{-7}W in ^{63}Ni source with the activity of 1 mCi (Section B. and (2)).

$$Pout = V_{oc}(v) \times I_{sc} \times FF \quad (10)$$

also, V_{OC} and J_{SC} are open-circuit voltage and short-circuit current, respectively. In I-V curve, V_{OC} is the voltage, at which the current is 0, and J_{SC} is the current, at which the voltage is 0. FF is the fill factor calculated by (11).

$$FF = \frac{P_{max}}{V_{OC}\, I_{SC}} \quad (11)$$

In this relation, P_{max} is the maximum value of (I_{sc}×V_{oc}) which can be read from power diagram.

Figs. 5 and 6 illustrate I-V and power diagrams of the structure designed based on Schottky junction between Pt and ZnO in non-porous and porous states, respectively. I-V curve entered the fourth region after irradiation, indicating the power is negative and is generated in the device. Short-circuit current, open-circuit voltage, FF, and efficiency of the non-porous structure are obtained as 12.36 nA, 1.115 V, 82.5%, and 11.37%, respectively. However, after creating pores on the surface exposed to radiation, these values increased to 37.5 nA, 1.23 V, 76.5%, and 35.5%, respectively. As can be seen, the short circuit current is considerably increased when ZnO nanowires are created on the surface. It shows that the number of the trapped beta particles is increased by using porous structure.

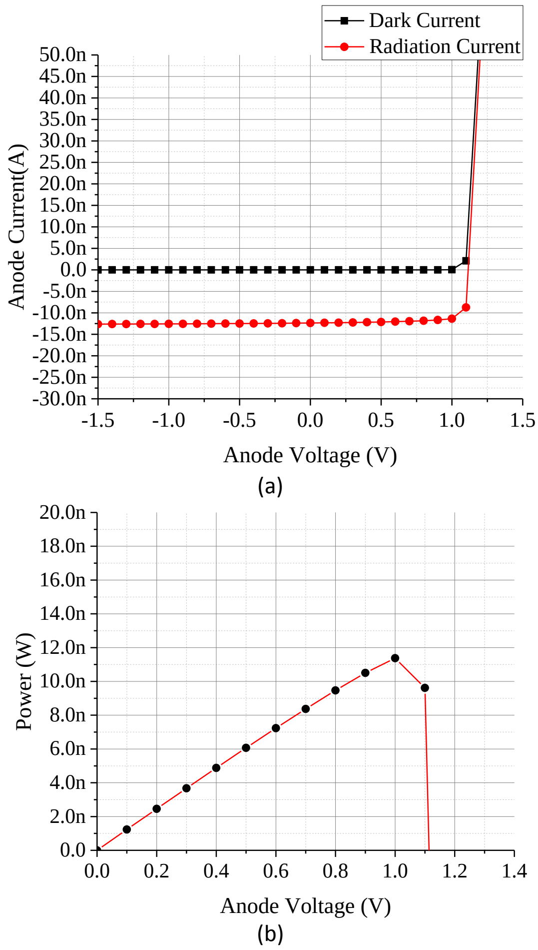

Fig. 5: a) I-V curve of non-porous structure in dark and radiation modes; b) Power diagram in terms of voltage for non-porous structure.

According to the formula below the short -circuit current is equal to [70] (12)

$$J_{sc} = N_e(\#pairs/s).1.6 \times 10^{18}(\frac{C}{pair}) \quad (12)$$

N_e shows the number of electron- hole pairs created per second in the depletion zone. Given the activity of the source of isotope and the comparison of structures in Table 4, this structure produces a suitable electron- hole pair. The VOC also has a semiconductor energy band.
Due to the semiconductor of zinc oxide on the maximum voltage of the open circuit of cell is 1.23 V, which has a good value compared to Activity and Table 4. The closer

the JSC times the VOC to Pmax, the fill factor is close to one and the structure is more suitable for an energy conversion cell.

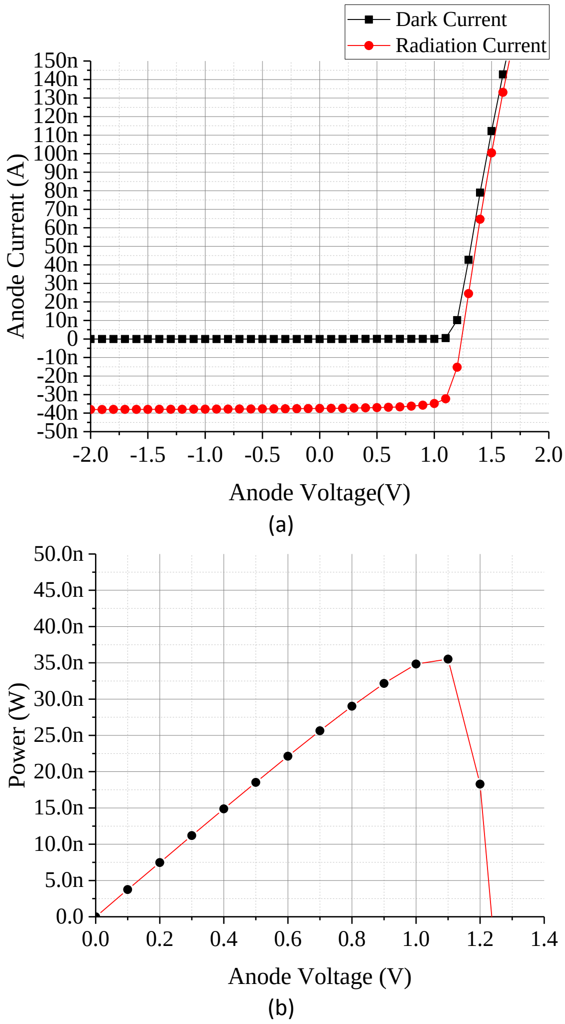

Fig. 6: a) I-V curve of porous structure in dark and radiation modes; b) Power diagram in terms of voltage for porous structure.

Table 3: Obtained results for different metals at Schottky junction with ZnO

Metal	short-circuit current(J_{SC})	open-circuit voltage (V_{OC})	Schottky barrier height	Maximum power (P_{max})
Pt	37.5 nA	1.237 V	1.54 ev	35.5 nW
Ir	37.1 nA	0.847 V	1.15 ev	22.9 nW
Ni	37 nA	0.74 V	1.05 ev	19.66 nW
Pd	36.9 nA	0.71 V	1.02 ev	18.54 nW
Au	36.8 nA	0.69 V	1 ev	17.26 nW

Comparison and Discussion

As shown in Table 2, various metals were investigated to form a Schottky junction with ZnO semiconductor in the porous structure, and the corresponding results are listed in Table 3. The short-circuit current values for Pt, Ir, Ni, Pd, and Au in the Schottky junction with ZnO were calculated as 37.5, 37.1, 37, 36.9, and 36.8 nA, respectively.

Additionally, their corresponding open-circuit voltage values were obtained as 1.237, 0.847, 0.74, 0.71, and 0.69 V, respectively. The results indicated Pt is the most suitable metal for forming Schottky junction in the structure due to its large work function. Fig. 7 illustrates the I-V curve in the radiation mode and the power diagram for different metals. To facilitate a comprehensive comparison of the cell results with other relevant published studies, Table 4 provides a summary of the findings from various betavoltaic cell investigations. The aim is to compile the characteristics of different betacells based on 63Ni. Generally, betacells utilizing wide bandgap materials tend to exhibit higher efficiency. Structures incorporating nanowires or porosity demonstrate improved short circuit current. This comparison highlights the superior performance of porous structures in betavoltaic battery outputs compared to non-porous structures.

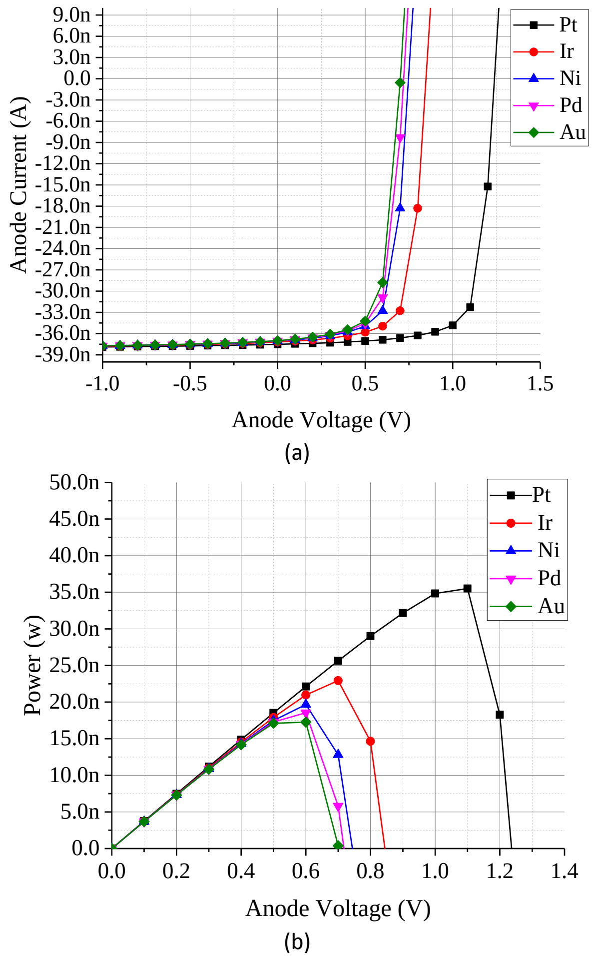

Fig. 7: a) I-V curve in radiation mode for different metals; b) Power diagram in terms of voltage for different metals.

Table 4: Comparing the obtained results with those of other studies

Reference	Efficiency	J_{SC}	V_{OC}	Junction	Semiconductor	Porous or Nanowires	Activity	Radiation source
This work	35.5%	37.5 nA	1.237 V	Schottky	Zno	yes	1 mci	^{63}Ni
[14]	4.4%	5.8 nA	2.1 V	p-i-n	GaN	no	-	^{63}Ni
[15]	1.5%	0.21 nA	0.95 V	p-i-n	SiC	no	0.16 mci	^{63}Ni
[18]	0.011%	1.2 nA/cm^2	0.3 V	p-i-n	GaN	no	50 mci	^{63}Ni
[21]	2.25%	7.5 nA	0.53 V	Schottky	GaN	no	1 mci	^{63}Ni
[33]	4.94%	573.3 nA	0.253 V	p-n	Si	no	100 mci	^{63}Ni
[47]	7.30%	12.43 nA	1.54 V	Schottky	TiO2	yes	8 mci	^{63}Ni
[50]	7.58%	0.1 uA	2.6 V	p-n	Zno	no	0.101 ci	^{63}Ni
[68]	2.85%	-	1.16 V	Schottky	GaN	no	-	^{63}Ni
[69]	4%	4.5 pA	0.69 V	p-i-n	GaInP	no	-	^{63}Ni
[71]	0.15%	13 nA/cm^2	6.5 mV	p-n	Si-SWNT	yes	3.3 mci	^{63}Ni
[72]	27.4 %	0.6 uA	4.9 v	p-n	Al0.7Ga0.3N	no	-	^{63}Ni
[73]	13.4%	16.36 nA	3.16 v	p-n	GaN	no	1 mci	^{147}Pm

Conclusion

This study explored a betavoltaic device with a porous structure based on a Schottky junction between Pt and ZnO semiconductor. The radioactive source employed was 63Ni, with a half-life of 100.2 years, an average energy of 17 keV, and an activity of 1 mCi, simulated using C++ code. The creation of pores effectively increased the contact surface area and trapped beta beams, resulting in improved performance metrics. The designed structure exhibited a short circuit current, open-circuit voltage, fill factor (FF), and efficiency of 37.5 nA, 1.237 V, 76.5%, and 35.5%, respectively.

Author Contributions

M. Amirmazlaghani designed the experiments. A. Ebadiyan performed the simulations. M. Amirmazlaghani, A. Ebadiyan and A. Shokri interpreted the results and wrote the manuscript. N. Darestani Farahani helped in validating and revising the manuscript.ach author role in the research participation must be mentioned clearly.

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Conflict of Interest

The authors declare no potential conflict of interest regarding the publication of this work. In addition, the ethical issues including plagiarism, informed consent, misconduct, data fabrication and, or falsification, double publication and, or submission, and redundancy have been completely witnessed by the authors.

Abbreviations

TCAD	Technology Computer-Aided Design
FF	Fill Factor

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